# PATENT ABSTRACTS OF JAPAN

(11)Publication number:

2002-134122

(43) Date of publication of application: 10.05.2002

(51)Int.CI.

H01M 4/90

B01J 27/186

B01.I 27/199

H01M 8/22

(21)Application number: 2000-327710

(22)Date of filing:

(71)Applicant: OTOMO JUNICHIRO

ON KELIIYO EGUCHI KOICHI

26.10.2000

(72)Inventor:

OTOMO JUNICHIRO

ON KELJIYO EGUCHI KOICHI

TAKAHASHI HIROSHI

TAKAHASHI HIROSHI

# (54) FUEL ELECTRODE MATERIAL FOR METHANOL FUEL CELL, METHANOL FUEL CELL, AND MANUFACTURING METHOD OF THEM

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a catalyst or a material for a methanol fuel electrode of a low price. direct type methanol fuel cell, the methanol fuel electrode having a catalyst for the material, and/or provide the

direct type methanol fuel fell having the catalyst or the material. SOLUTION: The methanol fuel electrode material for the direct type methanol fuel cell has a heteropoly acid comprising hydrogen, oxygen, phosphorus, and a first metal, such as H3PW12O40, a second metal and/or carbon.

1 of 1 8/15/2008 1:23 PM

## Disclaimer:

This English translation is produced by machine translation and may contain errors. The JPO, the INPIT, and those who drafted this document in the original language are not responsible for the result of the translation.

#### Notes:

1. Untranslatable words are replaced with asterisks (\*\*\*\*).

2. Texts in the figures are not translated and shown as it is.

### Translated: 03:29:30 JST 08/19/2008

Dictionary: Last updated 08/08/2008 / Priority: 1. Chemistry / 2. Mechanical engineering

#### CLAIM + DETAILED DESCRIPTION

## [Claim(s)]

[Claim 1] The methanol fuel electrode ingredient for direct type methanol fuel cells which has a heteropolyacid, the 1st metal, and/or carbon.

[Claim 2] Said ingredient is an ingredient according to claim 1 which has a heteropolyacid, the 1st metal, and carbon.

Claim 3] The ingredient according to claim 1 or 2 chosen from the group which said 1st metal becomes from platinum, ruthenium, platinum and the alloy of ruthenium, nickel, copper, iron, manganese, Cobalt, palladium, and caesium.

[Claim 4] 1st element; as which the architecture element is chosen from the group which said heteropolyacid becomes from a phosphorus, silicon, and germanium — the — the ingredient of Claim 1 which has element; hydrogen; of two, and oxygen - Claim 3 given in any 1 term.

Tryctoger, or two, and oxygen-claim 3 given many 1 from:
[Claim 5] The ingredient according to claim 4 chosen from the group which said 2nd element becomes from molybdenum, tungsten, vanadium, niobium, and a tantalum.

[Claim 6] Said heteropolyacid is an ingredient according to claim 4 or 5 which is one or more sorts chosen from the group which the 3rd element replaces said a part of hydrogen, and this 3rd element becomes from

platinum, nickel, copper, iron, caesium, manganese, Cobalt, and palladium. [Claim 7] The ingredient according to claim 6 said whose 3rd element is two or more sorts and whose 2nd kind the 1st kind of two or more sorts of inside is chosen from the group which consists of platinum, nickel, copper, and iron, and is caesium.

[Claim 8] Claim 1 said whose heteropolyacid is HIPMmOn (M shows the 2nd metal among a formula and I, m, and n are one or more integers) - the incredient of Claim 7 given in any 1 term.

[Claim 9] Claim 1 said whose heteropolyacid is HqPWrOs (the inside of a formula, q, r, and s are one or more

integers) - the ingredient of Claim 8 given in any 1 term.

[Claim 10] Claim 1 said whose heteropolyacid is H3PW12O40 - the ingredient of Claim 9 given in any 1 term.

[Claim 11] The fuel cell which is a fuel cell which has the electrolyte inserted into a cathode pole, an anode

[Claim 11] The fuel cell which is a fuel cell which has the electrolyte inserted into a cathode pole, an anode pole, and these two poles, and is characterized by said anode pole having a heteropolyacid, the 1st metal, and/or carbon.

[Claim 12] Said electrolyte is a fuel cell according to claim 11 which has a heteropolyacid, the 1st metal, and carbon.

[Claim 13] The fuel cell according to claim 11 or 12 with which said 1st metal is chosen from the group which consists of platinum, ruthenium, platinum and the alloy of ruthenium, nickel, copper, iron, manganese, Cobalt, palladium, and caesium.

[Claim 14] Ist element; as which the architecture element is chosen from the group which said heteropolyacid becomes from a phosphorus, silicon, and germanium – the – the fuel cell of Claim 11 which has element; hydrogen; of two, and oxygen – Claim 13 given in any 1 term.

[Claim 15] The fuel cell according to claim 12 with which said 2nd element is chosen from the group which consists of molybdenum, tungsten, vanadium, niobium, and a tantalum.

[Claim 16] Said heteropolyacid is an ingredient according to claim 14 or 15 which is one or more sorts chosen from the group which the 3rd element replaces said a part of hydrogen, and this 3rd element becomes from platinum, nickel, copper, iron, caesium, manganese, Cobalt, and palladium.

[Claim 17] The ingredient according to claim 16 said whose 3rd element is two or more sorts and whose 2nd kind the 1st kind of two or more sorts of inside is chosen from the group which consists of platinum, nickel.

more integers) - the fuel cell of Claim 18 given in any 1 term.

copper, and iron, and is caesium.

[Claim 18] Claim 11 said whose heteropolyacid is HIPMmOn (M shows the 2nd metal among a formula and I. m, and n are one or more integers) - the fuel cell of Claim 17 given in any 1 term. [Claim 19] Claim 11 said whose heteropolyacid is HqPWrOs (the inside of a formula, q, r, and s are one or

[Claim 20] Claim 11 said whose heteropolyacid is H3PW12O40 - the fuel cell of Claim 19 given in any 1 term. [Claim 21] Claim 11 said whose fuel cell is a direct type methanol solid polymer fuel cell - the fuel cell of Claim 20 given in any 1 term. [Claim 22] It is the manufacture method of the methanol fuel electrode ingredient for direct type methanol fuel

cells of having a heteropolyacid, the 1st metal, and/or carbon. The above-mentioned method of having the process which dissolves said heteropolyacid in a heteropolyacid-solubility solvent and obtains a solution, the process which suspends the 1st metal and/or carbon uniformly in this solution, and obtains suspension, and the process which dries this suspension and obtains said ingredient.

[Claim 23] Said ingredient is a method according to claim 22 of having a heteropolyacid, the 1st metal, and carbon.

[Claim 24] The method according to claim 22 or 23 by which said 1st metal is chosen from the group which consists of platinum, ruthenium, platinum and the alloy of ruthenium, nickel, copper, iron, manganese, Cobalt, palladium, and caesium.

[Claim 25] 1st element; as which the architecture element is chosen from the group which said heteropolyacid becomes from a phosphorus, silicon, and germanium -- the -- the method of Claim 22 which has element; hydrogen; of two, and oxygen - Claim 24 given in any 1 term.

[Claim 26] The method according to claim 25 by which said 2nd element is chosen from the group which consists of molybdenum, tungsten, vanadium, niobium, and a tantalum.

IClaim 271 Said heteropolyacid is an ingredient according to claim 25 or 26 which is one or more sorts chosen from the group which the 3rd element replaces said a part of hydrogen, and this 3rd element becomes from platinum, nickel, copper, iron, caesium, manganese, Cobalt, and palladium.

[Claim 28] The ingredient according to claim 27 said whose 3rd element is two or more sorts and whose 2nd kind the 1st kind of two or more sorts of inside is chosen from the group which consists of platinum, nickel, copper, and iron, and is caesium.

[Claim 29] Claim 22 - the method of Claim 28 given in any 1 term said heteropolyacid is HIPMmOn (M shows the 2nd metal among a formula and I, m, and n are one or more integers).

[Claim 30] Claim 22 - the method of Claim 29 given in any 1 term said heteropolyacid is HqPWrOs (the inside of a formula, q, r, and s are one or more integers).

IClaim 311 Claim 22 - the method of Claim 30 given in any 1 term said heteropolyacid is H3PW12O40.

[Claim 32] It is the fuel cell which has the electrolyte inserted into a cathode pole, an anode pole, and these two poles, and said anode pole is the manufacture method of a fuel cell of having a heteropolyacid, the 1st metal, and/or carbon. The process which dissolves said heteropolyacid in a heteropolyacid-solubility solvent and obtains a solution, The above-mentioned method of having the process which suspends the 1st metal and/or carbon uniformly in this solution, and obtains suspension, the process which applies this suspension to a base material, dries, and obtains said cathode pole, and the process at which this cathode pole and said anode pole are stuck as said electrolyte is inserted into.

[Claim 33] Said cathode pole is a method according to claim 32 of having a heteropolyacid, the 1st metal, and carbon.

IClaim 341 The method according to claim 32 or 33 by which said 1st metal is chosen from the group which consists of platinum, ruthenium, platinum and the alloy of ruthenium, nickel, copper, iron, manganese, Cobalt, palladium, and caesium.

[Claim 35] 1st element; as which the architecture element is chosen from the group which said heteropolyacid becomes from a phosphorus, silicon, and germanium -- the -- the method of Claim 32 which has element: hydrogen; of two, and oxygen - Claim 34 given in any 1 term.

[Claim 36] The method according to claim 35 by which said 2nd element is chosen from the group which consists of molybdenum, tungsten, vanadium, niobium, and a tantalum.

[Claim 37] Said heteropolyacid is an ingredient according to claim 35 or 36 which is one or more sorts chosen from the group which the 3rd element replaces said a part of hydrogen, and this 3rd element becomes from platinum, nickel, copper, iron, caesium, manganese, Cobalt, and palladium. [Claim 38] The ingredient according to claim 37 said whose 3rd element is two or more sorts and whose 2nd

kind the 1st kind of two or more sorts of inside is chosen from the group which consists of platinum, nickel, copper, and iron, and is caesium.

[Claim 39] Claim 32 - the method of Claim 38 given in any 1 term said heteropolyacid is HIPMmOn (M shows the 2nd metal among a formula and I, m, and n are one or more integers).

[Claim 40] Claim 32 - the method of Claim 39 given in any 1 term said heteropolyacid is HqPWrOs (the inside of a formula, q, r, and s are one or more integers).

[Claim 41] Claim 32 - the method of Claim 39 given in any 1 term said heteropolyacid is H3PW12O40.

# [Detailed Description of the Invention]

# [0001]

[Field of the Invention] This invention relates to the methanol fuel electrode (anode pole) ingredient for direct type methanol fuel cells. In particular, it is comparatively inexpensive and is related with the catalyst for methanol fuel electrode (anode pole) ingredients which shows high activity. Moreover, this invention relates to the manufacture method of these ingredients or a catalyst, a fuel cell, and its manufacture method.

## [0002]

[Description of the Prior Art] As a technique which reduces emission of the carbon dioxide which has caused the global warming issue, the fuel cell attracts attention. A fuel cell can be classified into some classified into some classified according to the difference in the electrolyte used, and has a phosphoric acid type (AFC), a meltisnge carbonate type (MCFC), a solid polymer type (PEFC), a solid acid ghost type (SOFC), etc. by it. In this, since PEFC can be operated at the low temperature in ordinary temperature -100 degree C, that utilization is expected, but PEFC which uses hydrogen as that fuel directly has a problem in the point of the storage condition of hydrogen, and the point that the volume density of energy is small.

[0003] The trouble produced by the method of using hydrogen directly is suppressed, and the method of using the hydrogen reformed on the other hand from methanol which is a liquid fuel is a leading method. Especially the direct methanol fuel cell (DMFC, Direct Methanol Fuel Cell) to which methanol is made to react on a direct electrode among the methods of using methanol attracts attention. That is, since a refining machine is unnecessary and small size and the weight saving are possible, DMFC is expected as portable power sources, such as a source for automobiles of electric power, and a cellular phone.

[0004] The electrode process of DMFC is as follows. That is, in a methanol fuel electrode (anode pole), it is CH3 OH+H2 O->CO2+6H++6e-, and is O2+4H++4e->2H2O in an air pole (cathode pole). In the abovementioned methanol fuel electrode, the reaction is promoted according to the catalyst, especially the catalyst which used Pt. Although Pt of the activity over the dissociative-adsorption process of methanol is high, the oxidation activity of intermediate products, such as CO generated in a methanol oxidation process and/or CHO, is low.

[0005] therefore, the duality which supported Pt to alloy [ with aPt, other metals, for example, Ru, Os Ir, and/or Sn ]; and b oxide, for example, Zro2, Tio2, Nb2O5, Ta2O5, WO3, etc. in order to promote the oxidation removal of an intermediate product – catalyst; etc. is tried. In these, since Ru promotes adsorption of water and OH is generated, the Pt-Ru catalyst has the highest activity.

#### 1000061

[Problem to be solved by the invention] However, in the present stage, engine performance for utilization even with a sufficient Pt-Ru catalyst which shows the highest activity is not obtained. Moreover, the both sides of Pt and Ru are expensive. [0007]

[Means for solving problem] Then, the object of this invention is to solve the above-mentioned technical problem. Specifically, the object of this invention is to offer the methanol fuel electrode which has a very inexpensive catalyst for methanol fuel electrodes, and this catalyst, and/or the direct type methanol fuel cell which has this catalyst.

[0008] Moreover, the catalyst for methanol fuel electrodes or ingredient which is [as for the object of this invention] comparable as a Pt-Ru catalyst other than the above-mentioned object in addition to the above mentioned object, or has the engine performance beyond it, It is in offering the direct type methanol fuel cell which has the methanol fuel electrode which has this catalyst or an ingredient and/or this catalyst, or an ingredient. Furthermore, in addition to the above-mentioned object, the object of this invention is to offer the manufacture method of the above-mentioned catalyst or an ingredient and the manufacture method of a fuel

cell other than the above-mentioned object.

[0009] This invention persons found out wholeheartedly that the above-mentioned technical problem was solvable by using the heteropolyacid which has the both sides of proton conductivity and the oxidation force, and the electronic conduction nature under reducing atmosphere as a result of analyses. That is, the following invention was found out

[0010] <1> Methanol fuel electrode ingredient for direct type methanol fuel cells which has a heteropolyacid, the 1st metal. and/or carbon.

<2> In the above <1>, the thing of an ingredient is [ having a heteropolyacid, the 1st metal, and carbon ] good. [0011] <3> In the above <1> or <2>, it is good to choose the 1st metal from the group which consists of platinum, ruthenium, platinum and the alloy of ruthenium, nickel, copper, iron, manganese, Cobalt, palladium, and caesium.

<4> 1st element, as which a heteropolyacid is chosen from the group which the architecture element becomes from a phosphorus, silicon, and germanium in the above <1> or <2> — the — having element; hydrogen; of two and oxygen has a good thing.

[0012] <5> In the above <4>, it is good to choose the 2nd element from the group which consists of molybdenum, tungsten, vanadium, niobium, and a tantalum.

<6> As for a heteropolyacid, in the above <4> or <5>, it is good that they are one or more sorts chosen from the group which the 3rd element replaces said a part of hydrogen, and this 3rd element becomes from platinum, nickel, copper, iron, caesium, manganese, Cobalt, and palladium.

[0013] <7> In the above <6>, the 3rd element is two or more sorts, the 1st kind of two or more sorts of inside is chosen from the group which consists of platinum, nickel, copper, and iron, and it is good that the 2nd kind is caesium.

<8> In either of above-mentioned <1>-<7>, it is good that a heteropolyacid is HIPMmOn (M shows the 2nd metal among a formula and I, m, and n are one or more integers).

[0014] <9> In either of above-mentioned <1>- <8>, it is good that a heteropolyacid is HqPWrOs (the inside of a formula, q, r, and s are one or more integers).

<10> In either of above-mentioned <1>- <9>, it is good that a heteropolyacid is H3PW12O40.

[0015] <11> Fuel cell which is a fuel cell which has the electrolyte inserted into a cathode pole, an anode pole, and these two poles, and is characterized by said anode pole having a heteropolyacid, the 1st metal, and/or carbon.

<12> In the above <11>, an electrolyte is good to have a heteropolyacid, the 1st metal, and carbon.

[0016] <13> In the above <11> or <12>, it is good to choose the 1st metal from the group which consists of platinum, ruthenium, platinum and the alloy of ruthenium, nickel, copper, iron, manganese, Cobalt, palladium, and caesium.

<14> 1st element, as which a heteropolyacid is chosen from the group which the architecture element becomes from a phosphorus, silicon, and germanium in either of above-mentioned <11> <13> -- the -- having element, hydrogen; of two and oxygen has a good thing.

[0017] <15> In the above <14>, it is good to choose the 2nd element from the group which consists of molybdenum, tungsten, vanadium, niobium, and a tantalum.

<16> As for a heteropolyacid, in the above <14> or <15>, it is good that they are one or more sorts chosen from the group which the 3rd element replaces said a part of hydrogen, and this 3rd element becomes from platinum, nickel, copper, iron, caesium, manganese, Cobalt, and palladium.

[0018] <17> In the above <16>, the 3rd element is two or more sorts, the 1st kind of two or more sorts of inside is chosen from the group which consists of platinum, nickel, copper, and iron, and it is good that the 2nd kind is caesium.

<18> In either of above-mentioned <11>- <17>, it is good that a heteropolyacid is HIPMmOn (M shows the 2nd metal among a formula and I, m, and n are one or more integers).

[0019] <19> In either of above-mentioned <11>- <18>, it is good that a heteropolyacid is HqPWrOs (the inside of a formula, q, r, and s are one or more integers).

<20> In either of above-mentioned <11>- <19>, it is good that a heteropolyacid is H3PW12O40.

21> In either of above-mentioned <11>-<20>, it is good that a fuel cell is a direct type methanol solid polymer fivel cell.

[0020] <22> It is the manufacture method of the methanol fuel electrode ingredient for direct type methanol fuel cells of having a heteropolyacid, the 1st metal, and/or carbon. The above-mentioned method of having the process which dissolves said heteropolyacid in a heteropolyacid-solubility solvent and obtains a solution, the

process which suspends the 1st metal and/or carbon uniformly in this solution, and obtains suspension, and the process which dries this suspension and obtains said ingredient.

[0021] <23> In the above <22>, the thing of an ingredient is [ having a heteropolyacid, the 1st metal, and carbon ] good.

<24> In the above <22> or <23>, it is good to choose the 1st metal from the group which consists of platinum, ruthenium, platinum and the alloy of ruthenium, nickel, copper, iron, manganese, Cobalt, palladium, and caesium.

[0022] <25> 1st element; as which a heteropolyacid is chosen from the group which the architecture element becomes from a phosphorus, silicon, and germanium in either of above-mentioned <22>- <24> — the — having element, hydrogen; of two and oxygen has a good thing. <26> In the above <25>, it is good to choose the 2nd element from the group which consists of molybdenum, tungsten, vanadium, niobium, and a tantalum. [0023] <27> As for a heteropolyacid, in the above <25> or <26>, it is good that they are one or more sorts chosen from the group which the 3rd element replaces said a part of hydrogen, and this 3rd element becomes from platinum, nickel, copper, iron, caesium, manganese, Cobalt, and palladium.

<28> In the above <27>, the 3rd element is two or more sorts, the 1st kind of two or more sorts of inside is chosen from the group which consists of platinum, nickel, copper, and iron, and it is good that the 2nd kind is caesium.

[0024] <29> In either of above-mentioned <22>- <28>, it is good that a heteropolyacid is HIPMmOn (M shows the 2nd metal among a formula and I, m, and n are one or more integers).

<30> In either of above-mentioned <22>- <29>, it is good that a heteropolyacid is HqPWrOs (the inside of a formula, q, r, and s are one or more integers).

<31> In either of above-mentioned <22> <30>, it is good that a heteropolyacid is H3PW12O40. [O025] <32> It is the fuel cell which has the electrolyte inserted into a cathode pole, an anode pole, and these two poles, and said anode pole is the manufacture method of a fuel cell of having a heteropolyacid, the 1st metal, and/or carbon. The process which dissolves said heteropolyacid in a heteropolyacid-solubility solvent and obtains a solution, The above-mentioned method of having the process which suspends the 1st metal and/or carbon uniformly in this solution, and obtains suspension, the process which applies this suspension to

a base material, dries, and obtains said cathode pole, and the process at which this cathode pole and said anode pole are stuck as said electrolyte is inserted into. 
[0026] <33> In the above <32>, a cathode pole is good to have a heteropolyacid, the 1st metal, and carbon. 
<34> In the above <32> or <33>, it is good to choose the 1st metal from the group which consists of platinum, ruthenium, platinum and the alloy of ruthenium, nickel, copper, iron, managenese, Cobalt, balladium, and

caesium. [0027] <a href="caesium">(0027] <a href="caesium">(35> 1st element</a>, as which a heteropolyacid is chosen from the group which the architecture element becomes from a phosphorus, silicon, and germanium in either of above-mentioned <a href="caesium">(32> <34> -- the -- having element, hydrogen; of two and oxygen has a good thing.

<36> In the above <35>, it is good to choose the 2nd metal from the group which an element becomes from molybdenum, tungsten, vanadium, niobium, and a tantalum.

[0028] <37> As for a heteropolyacid, in the above <35> or <36>, it is good that they are one or more sorts chosen from the group which the 3rd element replaces said a part of hydrogen, and this 3rd element becomes from platinum, nickel, copper, iron, caesium, manganese, Cobalt, and palladium.

<38> In the above <37>, the 3rd element is two or more sorts, the 1st kind of two or more sorts of inside is chosen from the group which consists of platinum, nickel, copper, and iron, and it is good that the 2nd kind is caesium.

[0029] <39> In either of above-mentioned <32>- <38>, it is good that a heteropolyacid is HIPMmOn (M shows the 2nd metal among a formula and I, m, and n are one or more integers).

<40> In either of above-mentioned <32>- <39>, it is good that a heteropolyacid is HqPWrOs (the inside of a formula, q, r, and s are one or more integers).

<41> In either of above-mentioned <32>- <40>, it is good that a heteropolyacid is H3PW12O40. [0030]

[Mode for carrying out the invention] This invention is hereafter explained more to a detail. The methanol fuel electrode ingredient for direct type methanol fuel cells of this invention has a heteropolyacid, the 1st metal, and/or carbon. That is, the ingredient of this invention has a heteropolyacid and the 1st metal, has a heteropolyacid and carbon, or has a heteropolyacid, the 1st metal, and carbon. The ingredient of this invention is good preferably to have a heteropolyacid, the 1st metal, and carbon.

[0031] (Heteropolyacid) The methanol fuel electrode ingredient of this invention contains a heteropolyacid. In this Description, a heteropolyacid means that in which the architecture element has hydrogen, oxygen, the 1st element, and the 2nd element metal. The 1st element is chosen from the group which consists of a phosphorus, silicon, and germanium. As for especially the 1st element, it is good that it is a phosphorus. [0032] Moreover, molybdenum (Mo) and tungsten (W), vanadium (V), niobium (Nb), and a tantalum (Ta) can be resolved as the 2nd element in a betagen break.

mentioned as the 2nd element in a heteropolyacid. [0033] Furthermore, a part of hydrogen of the heteropolyacid may be replaced by the 3rd element. The number of the 3rd element may be one, that of it may be two, or it may be three or more sorts. The 3rd element is good to be chosen out of the group which consists of platinum, nickel, copper, iron, caesium, manganese, Cobalt, and palladium. In addition, when the 3rd element is two or more sorts, the 1st kind is chosen from the group which consists of platinum, nickel, copper, and iron, and it is good that the 2nd kind is caesium. [0034] That is, the heteropolyacid which can be used in this invention can be expressed as HIE1mE2 m'On (E1 shows the 1st element among a formula, E2 shows the 2nd element, and I, m, m', and n are one or more integers), moreover, when a part of hydrogen is replaced by the 3rd element, a heteropolyacid can be expressed as HI'E3 m"E1mE2 m'On (a formula -- inside -- E -- one -- the -- one -- an element -- being shown --E -- two -- the -- two -- an element -- being shown -- E -- three -- three -- an element -- being shown -- I -- ' - m -- m -- '-- m -- " -- and -- n -- one or more integers -- it is). It is [among these] good that E1 is a phosphorus (P). Furthermore, it is good that E2 is tungsten (W) or molybdenum (Mo). That is, it is preferably [ often / that they are HqPWrOs (the inside of a formula, q, r, and s are one or more integers), or Hq'PMor'Os' (the inside of a formula, q', r', and s' are one or more integers) /, and ] good HqPWrOs and that it is H3PW12O40 more preferably.

[0035] Furthermore, a part of hydrogen (H) of H3PW12O40 can use what is replaced by the 3rd element as mentioned above. For example, H3-xE3xW12O40, H3-x-yE3xE4yPW12O40 (E3 and E4 show the 3rd above-mentioned element, and x and y show one or more integers), etc. can be mentioned. In addition, when using the 1st metal, the 3rd element of the above may be the same as this 1st metal.

[0036] (The 1st metal) [the 1st metal used for the methanol fuel electrode ingredient of this invention] Platinum (Pt), ruthenium (Ru), platinum, and alloy [of ruthenium];, and nickel (nickel), Copper (Cu) iron (Fe) (manganese Mn) Cobalt (Co), palladium (Pd), and caesium (Cs), And it is good to be chosen out of the group which consists of an alloy which consists or more of two of sorts of these, and it good to be chosen out of the group which consists of platinum (Pt), ruthenium (Ru), platinum, and an alloy of ruthenium especially. That is, the number of the 1st metal may be one, that of it may be two, or it may be three or more sorts.

[0037] (Other components) The methanol fuel electrode ingredient of this invention is good to have the ingredient which has proton conductivity other than the above, and has electronic conduction nature, for example, good to have Nafion etc. again.

[0038] (Amount of each component) As for the amount of each component used for the methanol fuel electrode ingredient of this invention mentioned above, it is good that they are the following ranges. That is, as for a heteropolyacid, it is preferably good among 100 weight % of methanol fuel electrode ingredients that it is 30 to 40 weight % ten to 70weight %.

[0039] When using carbon for the ingredient of this invention, as for the amount of this carbon, it is preferably good among 100 weight % of methanol fuel electrode ingredients that it is 15 to 20 weight % five to 40weight %. Moreover, the heteropolyacid in the ingredient of this invention: As for carbon (rate of a bulk density), it is good 1:4-14:1, and that it is 2:1 preferably.

[0040] When using the 1st metal for the ingredient of this invention, as for the amount of this 1st metal, it is preferably good among 100 weight % of methanol fuel electrode ingredients that it is 10 to 20 weight % five to 50weight %. Moreover, the heteropolyacid in the ingredient of this invention: As for the 1st metal (rate of a bulk density), it is good 1:5-14:1, and that it is 2:1 preferably.

[0041] (Process) The methanol fuel electrode ingredient of this invention can be prepared as follows, for example. That is, add a heteropolyacid to a heteropolyacid to solution is obtained. Subsequently, carbon and/or the 1st metal are added to this solution, and uniform suspension is obtained. It removes by drying a solvent from suspension, and a methanol fuel electrode ingredient is obtained. More specifically, water is used as the above-mentioned solvent. Moreover, a methanol fuel electrode ingredient can be obtained by applying the suspension obtained on the base material [\*\*\*\*/ carbon paper etc. /, for example ] for methanol fuel electrodes, and drying after that.

[0042] (Fuel cell) This invention also offers the fuel cell which has an above-mentioned methanol fuel electrode ingredient. A fuel cell has the electrolyte inserted into the anode pole, cathode pole, and two poles which have

an above-mentioned methanol fuel electrode ingredient.

[0043] As long as the anode pole of the fuel cell of this invention has an above-mentioned methanol fuel electrode ingredient, it may have other ingredients. This anode pole can be prepared as mentioned above. Moreover, as long as it has characteristics [\*\*\*\*] in a methanol fuel cell, a cathode pole may be better known than before, or may be developed in the future. Furthermore, as long as it has characteristics [\*\*\*\*], for example, proton conductivity, in a methanol fuel cell, an electrolyte may be better known than before, or may be developed in the future.

[0044] The fuel cell of this invention can be obtained by sticking the anode pole obtained above with a cathode pole so that an electrolyte may be inserted.

[Working example] This invention is hereafter explained more concretely using a work example. However, this example is only illustration and must not interpret as limiting this invention.

(Work example 1) Heteropolyacid H3PW12O40 and 0.10g were dissolved in 1.45g of water, and 12OH3PW40 aqueous solution was prepared. PVC (what supported Pt0.05g to 0.05g of carbon) 0.10g was put into this aqueous solution, it mixed uniformly, and suspension was obtained. Suspension was dried and 0.22g of H3PW12O40-Pt-C catalysts were acquired.

[0046] The paste which fully distributed the catalyst was obtained by putting [ three ] in 0.22g of acquired H3PW12O40-Pt-C catalysts in 1.96cm of 5%Nafion solutions prepared beforehand, and ultrasonicating them for 20 minutes. After applying obtained paste 60mg uniformly on circular carbon paper (14mm in diameter, and 0.30mm in thickness), it dried for 1 hour and the anode pole A-1 0.40mm in thickness and 14mm in diameter was obtained.

[0047] The cathode pole B-1 and the electrolyte C-1 were prepared independently [ the anode pole A-1 ]. The cathode pole B-1 was 0.4mm in thickness and the circle configuration 14mm in diameter which consist of Pt-Ru1.0 mg/cm2; carbon 1.0 mg/cm2; and Nafion1.7mg/cm2. Moreover, the electrolyte C-1 was the circle configuration Nafion with a thickness [89 micrometers of ], and a diameter of 40mm. The anode pole A-1 and the cathode pole B-1 have been arranged so that an electrolyte C-1 may be inserted, and it bonded by thermocompression for 5 minutes by 130 degrees C and 6.0MPa, and the electrode-electrolyte zygote X-1 was obtained.

[0048] (Comparative example 1) The anode pole A-2 which does not include a heteropolyacid was prepared like the anode pole A-1 of a work example 1. The electrode-electrolyte zygote X-2 was obtained using the cathode pole B-1, the same cathode pole as an electrolyte C-1, and electrolyte of a work example 1. In addition, the electrode-electrolyte zygote X-1 obtained by the work example 1 and the comparative example 1 and the component of X-2 were summarized in the following table 1. 10049]

[Table 1]

表 1. X-1及びX-2の成分

	,	触媒	Pt (mg/cm²)	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub> (mg/cm <sup>2</sup> )	C (mg/cm²)	Nafion (mg/cm²)
実施例 1	X-1	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub> & Pt/C	0.96	2. 1	1. 1	1. 6
H at al 1	Y - 2	P+/0	0 95	_	1 1	1 7

[0050] Moreover, the anode pole (fuel electrode) overvoltage and I-V characteristics as a fuel cell of X-1 and X-2 were measured. It carried out on condition of the following when measuring.

[0051] Temperature of 90 degrees C;

Anode pole (fuel electrode) gas: CH3OH A part for 10ml/, H2O 60mla part for /, Ar A part for 30ml/;

Cathode pole (air pole) gas: O2 A part for 100ml/(H2O bubbling).

[0052] The obtained result is shown in <u>drawing 1</u> and <u>drawing 2</u>. <u>Drawing 1</u> is a graph which shows the anode pole (fuel electrode) overvoltage of X-1 and X-2, and <u>drawing 2</u> is a graph which shows the I-V characteristics of X-1 and X-2. <u>Drawing 1</u> showed that the fuel cell of this invention using X-1 became small in a two or more 40 m/cm high current density field. That is, since the conducting route of the proton produced at the reaction by using a heteropolyacid increased and CO oxidation reaction of the heteropolyacid was promoted, the fuel cell of this invention using X-1 is considered to have obtained the above-mentioned result.

[0053] Moreover, as for the fuel cell of this invention using X-1, I-V characteristics showed that the grade of a voltage drop was suppressed and a highly efficient cell was obtained. [0054]

JP,2002-134122,A [CLAIM + DETAILED DESCRIPTION]

[Effect of the Invention] By this invention, the methanol fuel electrode which has a very inexpensive catalyst for methanol fuel electrodes and this catalyst, and/or the direct type methanol fuel cell which has this catalyst can be offered.

[0055] Moreover, the catalyst for methanol fuel electrodes or ingredient which is comparable as a Pt-Ru catalyst other than the above-mentioned effect, or has the engine performance beyond it in addition to the above-mentioned effect by this invention. The direct type methanol fuel cell which has the methanol fuel electrode which has this catalyst or an ingredient and/or this catalyst, or an ingredient can be offered. Furthermore, in addition to the above-mentioned effect, the manufacture method of the above-mentioned catalyst or an ingredient and the manufacture method of a fuel cell other than the above-mentioned effect can be offered by this invention.

[Translation done.]

8/18/2008 2:32 PM 8 of 8